Influence of disorder on magnetic properties and intrinsic anomalous Hall effect in epitaxial Co₂FeAl film

K. K. Meng^{1,*}, J. Miao¹, X. G. Xu¹, Y. Wu¹, J. H. Zhao² and Y. Jiang¹

¹ *School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, China*

² *State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China*

Abstract: We have investigated the influence of disorder on magnetic properties and intrinsic anomalous Hall effect (AHE) in epitaxial single-crystalline full-Heusler alloy Co₂FeAl. The magnetic properties are proved by x-ray absorption spectroscopy and x-ray magnetic circular dichroism measurements, and the magnetic moments of Co and Fe elements in both as-deposited and annealed Co₂FeAl films have been calculated. Using a proper scaling, we have also extracted the intrinsic anomalous Hall conductivity of the films. It is found that the AHC in the as-deposited films is thickness dependent, while in the annealed ones the value has not changed evidently. We ascribe this feature to the influence of chemical ordering by affecting the Fermi surface.

Keywords: Spintronics; Heusler alloy; Molecular-beam epitaxy; Magnetic properties

PACS: 85.75.-d; 75.47.Np; 81.15.Hi; 75.70.-i

* Authors to whom correspondence should be addressed:

kkmeng@ustb.edu.cn

Full-Heusler alloys belong to a group of ternary intermetallics with the stoichiometric composition X_2YZ ordered in an $L2_1$ -type structure, in which X and Y are transition metals and Z is usually a main group element. These ferromagnets (FM) possess a gap at Fermi level in the minority band but exhibit metallic behavior in the majority band, therefore the spin-polarization at the Fermi level is considered to be 100%. A high degree of spin polarization is interesting for various applications in magnetoelectronics. Recently, Co-based full-Heusler alloy in a chemical form of Co_2YZ are of particular interest due to their high Curie temperatures and high spin polarization. Some of these alloys including Co_2MnSi , Co_2FeAl and $Co_2Fe_xMn_{1-x}Si$ have been incorporated into magnetic tunnel junctions or current-perpendicular-to-plane giant magnetoresistance as ferromagnetic electrodes, achieving relatively high magnetoresistance. Therefore, the electronic band structure and magnetic properties of the Co-based Heusler compounds have been intensively investigated both theoretically and experimentally in recent years. Notably, the Co_2YZ can transform into disordered structures, such as $B2$ -type disorder due to mixing between Y and Z, $A2$ -type disorder due to complete mixing of Co, Y and Z. The magnetic properties will be affected by the mixing of the atoms, such as in Co_2CrAl alloy, the total magnetic moment decreases with increasing Co-Cr type disorder owing to the antiferromagnetic coupling of the anti-site Cr with the first nearest-neighbor ordinary-site Cr.

Furthermore, this kind of phase separation into two or more disordered phases is expected to result in reduced spin polarization because of the additional minority

density of states at the Fermi level. The topology of Fermi surface will correspondingly become complex, resulting in a modified spin-dependent transport properties such as anomalous Hall effect (AHE). The AHE which occurs in FM due to spin-orbit coupling (SOC) has resisted theoretical and experimental assaults for almost a century. It has various origins resulting from either the extrinsic impurity scattering processes or the intrinsic mechanism which can be interpreted as the Berry curvature of the occupied Bloch states. The possible mechanisms of AHE can be explored through giving a unified scaling describing the AHE resistivity ρ_{AH} in terms of the longitudinal resistivity ρ_{xx} . Karplus and Luttinger have proposed that the intrinsic AHE arises from the transverse velocity of Bloch electrons induced by SOC together with interband mixing, giving $\rho_{AH} \sim \rho_{xx}^2$. On the other hand, the extrinsic mechanisms including skew scattering and side jump come from the asymmetrical scattering of conduction electrons due to SOC, giving $\rho_{AH} \sim \rho_{xx}$ and $\rho_{AH} \sim \rho_{xx}^2$, respectively. Recent experimental work on $\text{Co}_2\text{FeSi}_{0.6}\text{Al}_{0.4}$ and $\text{Co}_2\text{FeGa}_{0.5}\text{Ge}_{0.5}$ films revealed that the intrinsic AHE can be significantly influenced by chemical ordering with affecting the topology of Fermi surface.

Working on epitaxial GaAs/Fe films, Tian *et al* have limited the scattering of electrons to two sources, one by interface roughness and another by phonons, with independent control on their strengths through the film thickness and sample temperature. They developed the experimental strategy neglecting the contribution of phonon skew scattering, giving the scaling:

$$\rho_{AH} = \alpha\rho_{xx} + \beta\rho_{xx}^2 + b\rho_{xx}^2 \quad (1)$$

where ρ_{xx0} is the residual resistivity induced by impurity scattering, ρ_{xx} denotes the longitudinal resistivity, α is the parameter of the skew scattering, β denotes the side jump, and b the intrinsic anomalous Hall conductivity (AHC). This scaling has been justified by theories and verified by experiments. Hou *et al* have investigated the AHE in epitaxial face-centered-cubic Co film grown by molecular beam epitaxy on MgO(001), and the intrinsic AHC is separated from the scattering related extrinsic contributions and is temperature independent. Ye *et al* have proposed a general experimental method to explore the unusual temperature dependence of AHE in Ni, which can extract the intrinsic AHC over the whole temperature range. As an attempt, we want to use this scaling to investigate the AHE in Co-based full-Heusler alloy.

In this paper, we have investigated the influence of disorder on magnetic properties and intrinsic AHE in epitaxial single-crystalline full-Heusler alloy Co_2FeAl with comparing the as-deposited films (in $A2$ structures) and the annealed ones (in $B2$ structures). The magnetic properties are proved by x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) measurements. The magnetic moments of Co and Fe elements have been calculated. Using Equation (1), we have also extracted the intrinsic AHC of the films. It is found that the AHC in the as-deposited films is thickness dependent, while in annealed ones the value is thickness independent. We ascribe this feature to the influence of chemical ordering by affecting the Fermi surface.

Co_2FeAl films with the thickness of $t=4, 8, 12$ nm were deposited on the more spatially isotropic $c(4\times4)$ reconstructed GaAs (001) surface by molecular-beam

epitaxy at room temperature. Reflection high energy electron diffraction was applied to in situ monitor the surface reconstruction during growth. After the deposition of the films, they were annealed at 400°C and 500°C to improve their crystal structure and the atomic order. The X-ray diffraction (XRD) profiles of the 12-nm-thick Co₂FeAl films as-deposited and annealed at various temperatures are shown in Figure. 1. Only the (004) Co₂FeAl peak and (002) and (004) GaAs substrate peaks were detected in the as-deposited films, indicating a *A2* order structure. The (002) Co₂FeAl peak appeared after annealing at 400°C and became slightly larger after annealing at 500°C, corresponding to a *B2* order structure.

In order to emphasize the effect from interface and localization, the thickness of CMA in this paper is in the range of 3~12 nm. Streaky RHEED patterns emerged during deposition as shown in Figure 1(a), indicating epitaxial growth. In order to protect the surfaces from oxidation, films were capped with 2 nm of aluminum. The *A2* structure was confirmed by the clear observation of (004) peak as discussed in our previous work [26]. The magnetic properties were investigated by a superconducting quantum interference device. The films were then patterned into Hall bars with a nominal length l of 2.5 mm and a width w of 0.2 mm using photolithography and ion-beam etching, and the transport properties were carried out in a physical property

measurement system (Quantum Design PPMS-9T system).

III. RESULTS AND DISCUSSION

Hysteresis loops of CMA films measured at 300 K under the external magnetic field along the [110] and [1-10] directions are shown in Fig. 1(b) and (c) respectively, and the magnetization is normalized to the saturation magnetization M_s . In-plane uniaxial magnetic anisotropy with easy axis parallel to the [110] direction is found. All the films have small coercive field, which is not evidently changed with varying thickness. In the [1-10] direction, discontinuities appear at the so-called split field which is a consequence of the superposition of the uniaxial and the fourfold anisotropy with the hard axis of the uniaxial anisotropy coinciding with an easy axis of the fourfold component [27]. The split field reduces from 200 to 10 Oe as increasing t from 3 to 12 nm. It is noted that the magnetic anisotropies of all the films should be attributable to the competition between uniaxial and the fourfold anisotropy. Considering the shape of hysteresis loops, both uniaxial anisotropy constant K_U and cubic anisotropy constant K_C were deduced using the method mentioned in Ref [27], and the results are shown in Fig. 2. It is found that K_C increases while K_U decreases as t increases, resulting smaller split fields. On the other word, the effect from the uniaxial anisotropy which mostly depends on the interface stress has become weak as increasing t .

For clarity we normalize the resistivity of all the films by the value at 300 K and plot the temperature dependence of $\rho_{xx}(T)/\rho_{xx}(300\text{ K})$ in Fig. 3(a). According to the Matthiessen rule, ρ_{xx} can be decomposed into the residual resistivity ρ_{xx0} at 5 K

originating from impurity scattering and ρ_{XXT} caused by finite temperature excitation by entities such as phonons and magnons. In low temperature regime, the resistivity in all the films has increased obviously as the temperature decreases, which can be ascribed to the localization. As shown in Fig. 3(b) the residual resistivity ρ_{XX0} increases with decreasing film thickness. The finite size effect on the resistivity is present in the thin film, and the interface scattering become significant. Because the interface scattering is inelastic, the electron mean free path is shortened by the presence of the interface and the resistivity is enhanced. The ρ_{AH} in all the films were obtained in the same way by subtracting the ordinary Hall component determined from a linear fit to the high-field region up to $\pm 4\text{T}$. Fig. 3 (c) demonstrates a representative set of hysteretic ρ_{AH} curves for the 12-nm-thick CMA film measured at various temperatures from 5 to 300 K. The temperature dependence of ρ_{AH} is plotted in Fig. 3(d), which shows an increasing trend with temperature. In the low temperature regime, the increase of ρ_{AH} is similar to the behavior of longitudinal resistivity, and the influence of localization on the AHE will also be investigated.

Considering that the conductivity of ultrathin CMA films is small enough, it is adequate to use the scaling given by Tian *et al* for describing the AHE in this system, which has been justified by theories and verified by experiments [22, 23]. Using this scaling, we can extract the intrinsic contribution from the total anomalous Hall conductivity. In Fig. 4(a), the ρ_{AH} is plotted versus the longitudinal resistivity square ρ_{XX}^2 . By linear fitting we found that for all the films these plots show good linearity from high temperature to low temperature, and the slope will be the intrinsic

anomalous Hall conductivity b shown in Equation (1). The intrinsic anomalous Hall conductivity b of different thicknesses obtained by the linear fitting in Fig. 4(a) are plotted together in Fig. 4(b). The intrinsic anomalous Hall conductivity noticeably decreases with thickness decreasing, which indicates that the size effect can change the Berry curvature contribution in CMA. The value of b for 12-nm-thick film has reached almost $1300 \Omega^{-1}\text{cm}^{-1}$. Kübler *et al* have computed Berry curvatures for a set of Heusler compounds using density functional calculations and the wave functions that they provide, and the anomalous Hall conductivity is obtained from the Berry curvatures [28]. Our value of b for 12-nm-thick film is close but smaller than the calculated bulk intrinsic anomalous Hall conductivity, while the intrinsic anomalous Hall conductivity for 3 nm film drops down to about $1100 \Omega^{-1}\text{cm}^{-1}$. It is found that the anomalous Hall conductivities can dramatically influenced by the band structure and the Fermi-surface topology. The smaller intrinsic anomalous Hall conductivity is most likely that the sample is in disordered A2 crystal structure and does not have the ideal Heusler L_{21} crystal structure. On the other hand, with varying the thickness of CMA, the chemical ordering will also be changed due to interface stress. Meanwhile, as the material approaching ultrathin films, the Berry curvature from bulk may gradually vanish. The intrinsic anomalous Hall conductivity, as the integral of all the Berry curvatures over the whole Brillouin zone, is supposed to decrease as observed in experiment. The influence of film thickness on intrinsic AHE needs further theoretical analysis in the future. On the other hand, to investigate the influence from localization in low temperature range, the skew scattering coefficient α and side-jump contribution

β are deduced by tuning thickness at different specific temperatures using the method in Ref [22]. Firstly, it should be pointed out that the Berry curvature in the CMA films could be temperature independent according to the nearly liner relationship of $\rho_{AH} \sim \rho_{XX}^2$. We now focus on the low temperature regime 5~100 K and $\rho_{AH} \ll \rho_{XX}$, taking the approximation $\sigma_{XX} \approx 1/\rho_{XX}$ and $\sigma_{AH} \approx -\rho_{AH} / \rho_{XX}^2$. Then the scaling Eq. (1) can be transformed to:

$$\frac{(-\sigma_{AH}(T)-b)\sigma_{xx0}^2}{\sigma_{xx}(T)^2} = \alpha\sigma_{xx0} + \beta \quad (2)$$

Using Eq. (2), α and β have been calculated and the results are shown in Fig. 4(c) and (d) respectively. In the range from 5 to 100 K, both α and β have not evidently changed, indicating both skew scattering and side jump contribution have negligible change. Therefore, the variance of α and β with temperature has a small impact on the total anomalous Hall conductivity, and the localization correction is weak.

IV. SUMMARY

In summary, we have investigated the thickness dependence of magnetic anisotropy and intrinsic AHE in single-crystalline full-Heusler alloy CMA. The magnetic anisotropy is the interplay of uniaxial and the fourfold anisotropy, and the corresponding anisotropy constants have been deduced. Considering the thickness of CMA is small, we ascribe it to the influence from interface stress. The AHE in CMA is found to be well described by the proper scaling $\rho_{AH} = \alpha\rho_{xx0} + \beta\rho_{xx0}^2 + b\rho_{xx}$. The intrinsic anomalous conductivity is found to be smaller than the calculated one and is thickness dependent, which is ascribed to the influence of chemical ordering by affecting the band structure and Fermi surface.

ACKNOWLEDGMENTS

This work was partially supported by the National Basic Research Program of China (2015CB921502), the National Science Foundation of China (Grant Nos. 61404125, 51371024, 51325101, 51271020).

References

- [1] R. A. de Groot, F. M. Müller, P. G. van Engen, and K. H. J. Buschow, Phys. Rev. Lett. **50**, 2024 (1983).
- [2] I. Galanakis, P. H. Dederichs, and N. Papanikolaou, Phys. Rev. B **66**, 174429 (2002).
- [3] S. Picozzi, A. Continenza and A. J. Freeman, Phys. Rev. B **66**, 094421 (2002).
- [4] G. H. Fecher, H. C. Kandpal, S. Wurmehl, J. Morais, H. J. Lin, H. J. Elmers, G. Schönhense and C. Felser, J. Phys.: Condens. Matter. **17**, 7237 (2005).
- [5] K. Özdoğan, E. Şaşıoğlu, B. Aktaş and I. Galanakis, Phys. Rev. B **74**, 172412 (2006)
- [6] W. H. Wang, M. Przybylski, W. Kuch, L. I. Chelaru, J. Wang, Y. F. Lu, J. Barthel, H. L. Meyerheim, and J. Kirschner, Phys. Rev. B **71**, 144416 (2005).
- [7] W. H. Wang, H. Sukegawa, R. Shan, S. Mitani, and K. Inomata, Appl. Phys. Lett. **95**, 182502 (2009).
- [8] Y. Sakuraba, J. Nakata, M. Oogane, Y. Ando, H. Kato, A. Sakuma, T. Miyazaki and H. Kubota, Appl. Phys. Lett. **88**, 022503 (2006).

- [9] Y. Sakuraba, M. Ueda, Y. Miura, K. Sato, S. Bosu, K. Saito, M. Shirai, T. J. Konno and K. Takanashi, Appl. Phys. Lett. **101**, 252408 (2012).
- [10] E. H. Hall, Philos. Mag. **10**, 301 (1880).
- [11] A. Hoffmann, IEEE Trans. Magn. **49**, 5172 (2013).
- [12] R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
- [13] G. Sundaram and Q. Niu, Phys. Rev. B **59**, 14915 (1999).
- [14] T. Jungwirth, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. **88**, 207208 (2002).
- [15] M. Onoda and N. Nagaosa, J. Phys. Soc. Jpn. **71**, 19 (2002).
- [16] J. Sinova, D. Culcer, Q. Niu, N. A. Sinitsyn, T. Jungwirth, and A. H. MacDonald, Phys. Rev. Lett. **92**, 126603 (2004).
- [17] D. Xiao, M.-C. Chang, and Q. Niu, Rev. Mod. Phys. **82**, 1959 (2010).
- [18] J. Smit, Physica (Amsterdam) **21**, 877 (1955).
- [19] L. Berger, Phys. Rev. B **2**, 4559 (1970).
- [20] A. Crepieux and P. Bruno, Phys. Rev. B **64**, 014416 (2001).
- [21] Y. Tian, L. Ye, and X. Jin, Phys. Rev. Lett. **103**, 087206 (2009).
- [22] L. Wu, K. Zhu, D. Yue, Y. Tian and X. F. Jin, Phys. Rev. B **93**, 214418 (2016).
- [23] D. Z. Hou, Y. F. Li, D. H. Wei, D. Tian, L. Wu and X. F. Jin, J. Phys.: Condens. Matter **24**, 482001 (2012).
- [24] P. He, L. Ma, Z. Shi, G. Y. Guo, J. G. Zheng, Y. Xin, and S. M. Zhou, Phys. Rev. Lett. **109**, 066402 (2012).
- [25] H. Scheider, E. V. Vidal, S. Chadov, G. H. Fecher, C. Felser, and G. Jakob, J. Magn. Mater. **322**, 579 (2010).

- [26] K. K. Meng, S. L. Wang, P. F. Xu, L. Chen, W. S. Yan, and J. H. Zhao, Appl. Phys. Lett. **97**, 232506 (2010).
- [27] M. Dumm, M. Zöfl, R. Moosbühler, M. Brockmann, T. Schmidt, and G. Bayreuther, J. Appl. Phys. **87**, 5457 (2000).
- [28] J. Kübler and C. Felser, Phys. Rev. B **85**, 012405 (2012).

Figure Captions

Fig. 1 (a) The RHEED pattern of CMA films. (b) Normalized in-plane hysteresis loops of CMA films with varying thickness, and the field is applied along $[110]$ direction. (c) Normalized in-plane hysteresis loops of CMA films with the field applied along $[1-10]$ direction.

Fig. 2 (a) Effective cubic magnetic anisotropy constant K_C and (b) effective uniaxial anisotropy constant K_U for CMA films with varying thickness.

Fig. 3 (a) The temperature dependence of the longitudinal resistivity ρ_{XX} for the six samples. The data are normalized by the values at 300 K. (B) The residual resistivity

ρ_{XX0} plotted against the film thickness. (c) ρ_{AH} -H curves at different temperatures for 12-nm-thick CMA film. (d) ρ_{AH} as the function of temperature for all the films.

Fig. 4 (a) ρ_{AH} vs ρ_{XX}^2 for different thicknesses of CMA films. (b) The thickness dependance of the intrinsic intrinsic anomalous Hall conductivity b . (c) and (d) skew scattering coefficient α and side-jump contribution β vs temperatures respectively.